

Electron spin relaxation by nuclei in semiconductor quantum dots

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Abstract

We have studied theoretically the electron spin relaxation in semiconductor quantum dots via interaction with nuclear spins. The relaxation is shown to be determined by three processes: (i) – the precession of the electron spin in the hyperfine field of the frozen fluctuation of the nuclear spins; (ii) – the precession of the nuclear spins in the hyperfine field of the electron; and (iii) – the precession of the nuclear spin in the dipole field of its nuclear neighbors. In external magnetic fields the relaxation of electron spins directed along the magnetic field is suppressed. Electron spins directed transverse to the magnetic field relax completely in a time on the order of the precession period of its spin in the field of the frozen fluctuation of the nuclear spins. Comparison with experiment shows that the hyperfine interaction with nuclei may be the dominant mechanism of electron spin relaxation in quantum dots.

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I. INTRODUCTION

The long electron spin dephasing time (hundred nanoseconds) recently reported [1] suggests using the spin of an electron localized in a quantum dot (QD) as the realization of a quantum bit, and electron doped quantum dots as the basic material for implementing a solid state based quantum computer [2]. Room temperature operation, which is usually a primary requirement in microelectronics, is of secondary importance for quantum computers relative to that of finding a material for their physical realization. Rather, operating now at low temperature, where the localized electron has a long spin coherence time, is an essential condition for quantum computing and spin memory storage. The dominant electron spin relaxation mechanism in bulk is connected with the spin-orbit interaction of carriers (see Ref. [3]), but is strongly suppressed for localized carriers [4]. Spin relaxation due to the electron hole exchange interaction plays an important role during the time of nonequilibrium carrier relaxation [5–7], but does not effect localized electron spin relaxation afterward. Dephasing of the electron in QD ground states via two phonon real transitions to, or virtual transitions through, excited states (Urbach or Raman two phonon processes [8]) is also suppressed at low temperatures [9]. As a result electron spin relaxation via interaction with nuclei becomes the dominant relaxation mechanism for localized electrons at low temperatures.

The interaction of localized electrons with nuclei was studied early on for electrons localized at donors in bulk (see for example [10,11]). There, the electron interacts with a large number of nuclei and feels the hyperfine magnetic field of the nuclei located in the region where electron is localized; this is also true for QD localized electrons. However, the correlation time of the electron-nucleus interaction of donor localized electrons is limited by the time of shallow donor ionization, and tunnel jumps between the donors, τ_c . Usually this time is much shorter than the period of the electron spin precession, ω_f^{-1} , in the hyperfine field of the nuclei. For quantum computation and spin storage, however, it is important to have a large value of τ_c , and we are interested in the opposite limit: $\omega_f \tau_c \gg 1$. This is realized in QDs at sufficiently low temperatures.

In this paper we consider the electron spin relaxation via its interaction with the spins of the nuclei in QDs in the absence and the presence of an external magnetic field. The process is facilitated by the disparity of the characteristic time scales of the three processes that determine the relaxation: the period of the electron precession in the frozen fluctuation of the hyperfine field of the nuclei, the period of the nuclear spin precession in the hyperfine field of the electron, and the nuclear spin relaxation time in the dipole-dipole field of its nuclear neighbors. Estimates of these time scales can be made for the case of GaAs, whose hyperfine constants are well known. For QDs containing 10^5 nuclei they are found to be: ~ 1 ns, ~ 1 μ s and ~ 100 μ s, respectively. Therefore, as a first step, we can describe the electron spin relaxation as a precession in the quasi-stationary frozen fluctuation of the hyperfine field of the nuclear spins. We can then examine the additional electron spin relaxation arising from the slowly varying nuclear hyperfine fields. The nuclear dipole-dipole interaction does not conserve the total nuclear spin; the third time scale provides a natural limit on the coherence of the electron-nuclei spin system. However many other relaxation mechanisms are important at this long time scale, therefore and we will not consider the effect of the nuclear dipole-dipole interactions on the electron spin relaxation. A spin dephasing time (T_2^*) calculated for an ensemble of QDs is in a good agreement with available experimental data.

The paper is organized as follows: The hyperfine interaction of an electron with the nuclei and the hyperfine nuclear magnetic fields acting on the electron are considered in section II. The electron spin relaxation times in the absence and in the presence of an external magnetic field are calculated in sections III and IV respectively. In section V we compare our theoretical results with experimental data.

II. HYPERFINE INTERACTION OF A LOCALIZED ELECTRON WITH NUCLEI. THE FROZEN FLUCTUATION OF THE NUCLEAR HYPERFINE FIELD IN QUANTUM DOTS.

The electron spin relaxation due to the nuclear spins is determined by their hyperfine Fermi contact interaction. The Hamiltonian of this interaction can be written [12]:

$$\hat{H}_{\text{cont}} = \frac{16\pi}{3} \mu_B \sum_j \frac{\mu_j}{I_j} (\hat{\mathbf{S}} \cdot \hat{\mathbf{I}}_j) \delta(\mathbf{r} - \mathbf{R}_j) , \quad (1)$$

where μ_B is the Bohr magneton, $\hat{\mathbf{S}}$ and \mathbf{r} are the spin and position of the electron, μ_j , $\hat{\mathbf{I}}_j$ and \mathbf{R}_j are the magnetic moment, spin and position of the j -th nucleus, and the sum goes over all the nuclei in the lattice. For localized electrons the distance between their energy levels is much larger than the energy of the hyperfine interaction with the nuclei. As a result this spin-spin interaction can be described by the Hamiltonian:

$$\hat{H}_{\text{hf}} = \frac{v_0}{2} \sum_j A^j |\psi(\mathbf{R}_j)|^2 \left(\hat{I}_z^j \hat{\sigma}_z + \hat{I}_x^j \hat{\sigma}_x + \hat{I}_y^j \hat{\sigma}_y \right) , \quad (2)$$

which is obtained from first order perturbation theory. Here v_0 is the volume of the unit cell, $\psi(\mathbf{R}_j)$ is the electron envelope wave function at the j -th nucleus, \hat{I}_α and $\hat{\sigma}_\alpha$ are the spin projections on the coordinate axes $\alpha = x, y, z$, and $A^j = (16\pi\mu_B\mu_j/3I^j) |u_c(\mathbf{R}_j)|^2$, where $u_c(\mathbf{R}_j)$ is the electron Bloch function at the nucleus. In GaAs, the sum of A^j over all the nuclei in the unit cell $A = \sum_j A^j \approx 90 \mu\text{eV}$ [13]. We can neglect the interaction of holes with the nuclei because the hole Bloch functions vanish at the nuclear positions. In addition, we neglect, for now, the nuclear dipole-dipole interactions, which do not conserve the total spin of the electron-nuclear system. They become important only at times longer than 10^{-4} s.

The effective nuclear hyperfine magnetic field, \mathbf{B}_N , acting on a localized electron spin can be obtained from Eq.2 taking the expectation of the Hamiltonian \hat{H}_{hf} over the ensemble of nuclear wave functions. It is the sum of contributions from a large number of nuclei:

$$\mathbf{B}_N = \frac{\nu_0}{\mu_B g_e} \langle \sum_j A^j |\psi(\mathbf{R}_j)|^2 \hat{\mathbf{I}}^j \rangle_N \quad (3)$$

where $\langle \dots \rangle_N$ denotes a quantum mechanical average over the ensemble of nuclear wave functions and g_e is the electron g -factor. We should note that the precession frequency of the electron in the hyperfine field of all the nuclei is much greater than the precession frequency of a nucleus in the hyperfine field of the electron. That is, the electron sees a snapshot of the "frozen fluctuation" of the nuclear field. The magnitude and direction of this field are randomly distributed, and described by a Gaussian probability density distribution function:

$$W(\mathbf{B}_N) = \frac{1}{\pi^{3/2} \Delta_B^3} \exp \left[-\frac{(\mathbf{B}_N)^2}{\Delta_B^2} \right], \quad (4)$$

where Δ_B is the dispersion of the nuclear hyperfine field distribution,

$$\Delta_B^2 = \frac{2}{3} \langle (\mathbf{B}_N)^2 \rangle = \frac{2}{3} \sum_j I^j (I^j + 1) (a_j)^2, \quad (5)$$

where

$$a_j = (v_0 / \mu_B g_e) A^j |\psi(\mathbf{R}_j)|^2 \quad (6)$$

is the magnetic field of a single nuclear spin acting on the electron, and we assumed that the nuclear spin directions are independent of each other. All nuclei in GaAs have the same spin $I^j = I = 3/2$. Replacing the sum over unit cells by an integration we obtain:

$$\Delta_B^2 = \frac{2I(I+1)}{3} \frac{\sum_j (A^j)^2}{(\mu_B g_e)^2} \frac{v_0}{V_L} = \frac{16I(I+1)}{3N_L} \frac{\sum_j (A^j)^2}{(\mu_B g_e)^2}, \quad (7)$$

where the sum in this equation goes over only those nuclei in a unit cell,

$$V_L = \left(\int d^3r \psi^4(\mathbf{r}) \right)^{-1} \text{ and } N_L = 8V_L / v_0 \quad (8)$$

is the number of nuclei in the volume, V_L , that effectively determine the electron precession frequency. In GaAs, the sum $\sum_j (A^j)^2 \approx 1.2 \cdot 10^{-3} \text{ meV}^2$. The volume, V_L , is on the order of the volume of the electron localization.

III. ELECTRON SPIN RELAXATION IN ZERO EXTERNAL MAGNETIC FIELD

A. Electron spin dephasing in the frozen fluctuations of the nuclear field

Let us consider an ensemble of identical QDs in which we simultaneously (at time $t = 0$) create electrons all having the same spin orientation (They can be created, for example, by circularly polarized light). The nuclear spins in the QDs of this ensemble are randomly oriented, the nuclear hyperfine fields in the dots differ from one another and, therefore, have a different effect on the initial electron spin, \mathbf{S}_0 , in each dot. We will consider the time dependence of the ensemble average electron spin relaxation for times small relative to the period of the nuclear precession in the hyperfine field of the electron. Each electron spin will be moving in the frozen fluctuation of the nuclear hyperfine magnetic field, \mathbf{B}_N , (see Eq.3) in its own QD. These fields, however, are randomly distributed among the dots of the ensemble. Therefore, even though each electron spin will precess in a coherent fashion in the frozen hyperfine field of its own dot, the ensemble average spin polarization will decrease.

The equation of motion of the spin \mathbf{S} in a fixed magnetic field, \mathbf{B} , is given by:

$$\begin{aligned} \mathbf{S}(t) = & (\mathbf{S}_0 \cdot \mathbf{n})\mathbf{n} + \{\mathbf{S}_0 - (\mathbf{S}_0 \cdot \mathbf{n})\mathbf{n}\} \cos \omega t \\ & + [\{\mathbf{S}_0 - (\mathbf{S}_0 \cdot \mathbf{n})\mathbf{n}\} \times \mathbf{n}] \sin \omega t , \end{aligned} \quad (9)$$

where \mathbf{S}_0 is the initial spin, $\mathbf{n} = \mathbf{B}/B$ is a unit vector in the direction of the magnetic field, and $\omega = \mu_B g_e B / \hbar$ is the Larmor frequency of the electron precession in this field. The equation also describes the coherent electron spin precession in a single QD due to the magnetic field, \mathbf{B}_N , of the frozen fluctuation of the nuclei ($\mathbf{n} = \mathbf{B}_N/B_N$, and $\omega = \mu_B g_e B_N / \hbar$). Averaging Eq.(9) over the magnetic field distribution of Eq.(4), we obtain the time dependence of the ensemble averaged electron spin polarization:

$$\langle \mathbf{S}(t) \rangle = \frac{\mathbf{S}_0}{3} \left\{ 1 + 2 \left[1 - 2 \left(\frac{t}{T_\Delta} \right)^2 \right] \exp \left[- \left(\frac{t}{T_\Delta} \right)^2 \right] \right\} . \quad (10)$$

The same time dependence describes the electron spin polarization of a single quantum dot averaged over a large number of measurements. Here

$$T_\Delta = \frac{\hbar}{\mu_B g_e \Delta_B} = \hbar \sqrt{\frac{3N_L}{16 \sum_j I^j (I^j + 1) (A^j)^2}} \quad (11)$$

is the ensemble dephasing time which arises from the random electron precession frequencies in the randomly distributed frozen fluctuation of the nuclear hyperfine field in the dots. This time is on the order of 1 ns for GaAs quantum dots with 10^5 nuclei. The spin dephasing time is proportional to $\sqrt{V_L}$. One can see that the average electron polarization relaxes to 10% of its original value after a time equal to the dephasing time and then increases to a steady state value of 33% of its initial polarization.

B. Electron spin dephasing as a result of variations of the nuclear field direction

A localized electron interacts with a large number of nuclei, $N_L \gg 1$. The interaction of the electron spin with a single nucleus is $\sqrt{N_L}$ times weaker than its interaction with the effective magnetic field of the frozen nuclear fluctuation. Changing the direction of a single nuclear spin only weakly perturbs the electron spin motion. The precession of the electron in the macroscopic fluctuation of the nuclear spins is $\sqrt{N_L}$ times faster than the precession of a nucleus in the hyperfine field of an electron, i.e., the nuclear precession period $T_N \sim T_\Delta \sqrt{N_L}$. Since an electron in a QD precesses so rapidly around the nuclear magnetic field \mathbf{B}_N , the nuclei only see the long time average of the hyperfine field of the electron, which is directed along \mathbf{B}_N . Note, that components of the nuclear field perpendicular to \mathbf{B}_N cancel each other out. Now, however, each nucleus, in turn, precesses about this direction with a different precession rate that is proportional to the square of the electron wave function at their respective nuclear positions. This variation in the precession rates leads to a non-vanishing slow time varying change in the frozen fluctuation of the hyperfine magnetic field of the nuclei, $\Delta \mathbf{B}_N \sim \sum_j (A^j |\psi(\mathbf{R}_j)|^2)^2 \langle \hat{\mathbf{I}}^j \times \mathbf{B}_N \rangle_N$, that is perpendicular to \mathbf{B}_N . These random changes in the nuclear magnetic field result in an additional relaxation of the electron spin polarization.

Equation (10) describes the time dependence of the electron spin relaxation in the frozen fluctuation of the nuclear hyperfine fields for times much less than T_N . To include the effect of the time dependent changes in the nuclear fields, we need to examine the ensemble average

of Eq.(9) at a much later time than is considered in Eq.(10). Consider the ensemble average of Eq.(9), but now at times for which the nuclear fields are time dependent. In addition, we average this quantity over a time interval large compared to the period of the electron precession but small compared to the time at which the ensemble average is taken. This leads to:

$$\langle \mathbf{S}(t) \rangle = \langle \mathbf{n}(t)(\mathbf{n}(t) \cdot \mathbf{S}(t)) \rangle = \left\langle \frac{\mathbf{B}_N(t)(\mathbf{B}_N(t) \cdot \mathbf{S}(t))}{B_N^2(t)} \right\rangle. \quad (12)$$

In this equation $(\mathbf{B}_N(t) \cdot \mathbf{S}(t))$ is the energy of the electron-nucleus spin system, which does not depend on time and is equal to $(\mathbf{B}_N(0) \cdot \mathbf{S}(0))$. Nor does B_N^2 change its value, because $d\mathbf{B}_N/dt \perp \mathbf{B}_N$. As a result,

$$\langle \mathbf{S}(t) \rangle = \left\langle \frac{\mathbf{B}_N(t)(\mathbf{B}_N(0) \cdot \mathbf{S}(0))}{B_N^2(0)} \right\rangle. \quad (13)$$

The rotational symmetry of the Hamiltonian Eq.2 leads to $\langle \mathbf{B}_N(t)_\alpha \mathbf{B}_N(0)_\beta \rangle = \delta_{\alpha,\beta} \langle \mathbf{B}_N(t)_x \mathbf{B}_N(0)_x \rangle = (\delta_{\alpha,\beta}/3) \langle (\mathbf{B}_N(t) \cdot \mathbf{B}_N(0)) \rangle$. This allows us to write Eq.13 as

$$\langle \mathbf{S}(t) \rangle = \left\langle \frac{(\mathbf{B}_N(t) \cdot \mathbf{B}_N(0))}{B_N^2(0)} \right\rangle \frac{\mathbf{S}(0)}{3}. \quad (14)$$

We see again that $\langle \mathbf{S}(t) \rangle$ is directed along $\mathbf{S}(0)$, which is the only physically defined direction in the ensemble. This equation describes the time dependence of the electron spin relaxation at times large compared to T_Δ and is limited only by the characteristic time scale of the dipole-dipole interactions. One can see that at times $T_\Delta < t < T_N$ Eq.(14) gives the same result as Eq.(10).

The variation of nuclear magnetic field direction in the time dependent correlation, in Eq.(14), is limited by the conservation of the total spin angular momentum of the electron-nucleus spin system, $\hat{\mathbf{F}} = \hat{\mathbf{S}} + \sum_j \hat{\mathbf{I}}^j$. The conservation of $\hat{\mathbf{F}}$ at times shorter than the nuclear dipole-dipole relaxation time follows from the fact that $\hat{\mathbf{F}}$ commutes with the Hamiltonian of the electron-nucleus spin system Eq.(2). The total nuclear spin is, then, effectively conserved since: $\mathbf{I}_\Sigma = \sum_j \mathbf{I}_j = \mathbf{F} - \mathbf{S} \approx \mathbf{F}$ because $F \gg S$ [14]. The latter follows from the fact that the dispersion of \mathbf{I}_Σ increases with the number of nuclei in each QD and the average value of $|\mathbf{I}_\Sigma| \gg 1$.

If the electron wave function were constant in the localization region and zero outside (the so called "box model" [15]) the nuclear magnetic field, \mathbf{B}_N , would be proportional to the total nuclei spin, \mathbf{I}_Σ , and would also be conserved. From Eq.(14) the nuclear spin precession would not then lead to any additional spin relaxation in this model, and $\langle \mathbf{S}(t) \rangle = \mathbf{S}_0/3$. However, in real QDs the amplitude of the electron wave function at the nuclei in the localization region does depend on their position. The nuclear field is not uniquely determined by the value and direction of the total nuclear spin; a distribution of values of \mathbf{B}_N are possible for the same value of \mathbf{I}_Σ . The total nuclear spin, \mathbf{I}_Σ , can be distributed in different ways among the nuclei as a result of their interaction with the electron.

The determination of the correlation, Eq.(14) at all times is beyond the scope of this paper. We will evaluate it only in the limit $t \gg T_N$, where the $\mathbf{B}_N(t)$ are randomly distributed. The joint distribution function of the nuclear fields and total nuclear spin can be written as the product of the total nuclear spin distribution function and the conditional probability distribution of nuclear fields given a certain value of the total nuclear spin:

$$W(\mathbf{B}_N, \mathbf{I}_\Sigma) = W_I(\mathbf{I}_\Sigma) w(\mathbf{B}_N | \mathbf{I}_\Sigma) . \quad (15)$$

Both of these latter distributions have a Gaussian form [16]:

$$W_I(\mathbf{I}_\Sigma) = \frac{1}{\pi^{3/2} \Delta_I^3} \exp \left[-\frac{(\mathbf{I}_\Sigma)^2}{\Delta_I^2} \right] , \quad (16)$$

and

$$w(\mathbf{B}_N | \mathbf{I}_\Sigma) = \frac{1}{\pi^{3/2} \Delta_B'^3} \exp \left[-\frac{(\mathbf{B}_N - \langle a \rangle \mathbf{I}_\Sigma)^2}{\Delta_B'^2} \right] , \quad (17)$$

where $\Delta_I^2 = 2NI(I+1)/3$ is the dispersion of the total nuclear spin distribution, and N is the number of nuclei which are in statistical equilibrium with the localized electron (see Appendix). This number is approximately equal to the number of nuclei in the dot. The conditional distribution of the random hyperfine fields, given a value \mathbf{I}_Σ for the total nuclear spin, is shifted from zero to the field due to the weighed total nuclei spin, $\langle a \rangle \mathbf{I}_\Sigma$, where $\langle a \rangle = (\sum_j^N a_j)/N = A/(\mu_B g_e N)$. The dispersion of this distribution, $\Delta_B'^2 = \Delta_B^2 - \langle a \rangle^2 \Delta_I^2$, is

determined by integrating Eq.17 with distribution Eq.(16) over all \mathbf{I}_Σ and comparing it with Eq.(4). Clearly $\Delta_B'^2 < \Delta_B^2$, because the latter also includes the dispersion of the random distribution of the total nuclear spin.

In general, to calculate the correlation between the direction of the hyperfine nuclear field at two times, $\mathbf{n}_0 = \mathbf{B}_N(0)/B_N(0)$, and $\mathbf{n}(t) = \mathbf{B}_N(t)/B_N(t)$, (see Eq.(14)), we have to find the angular distribution of the magnetic field $\mathbf{B}_N(t)$ given a certain value both of the total nuclear spin \mathbf{I} and the magnetic field magnitude, $B_N = |\mathbf{B}_N|$. This conditional distribution function is written:

$$F(\mathbf{n}|B_N, \mathbf{I}) = \frac{\exp \left[-(B_N \mathbf{n} - \langle a \rangle \mathbf{I})^2 / \Delta_B'^2 \right]}{\int d\Omega(\mathbf{n}) \exp \left[-(B_N \mathbf{n} - \langle a \rangle \mathbf{I})^2 / \Delta_B'^2 \right]}, \quad (18)$$

Using distributions of Eqs.(4,16,18) we find the correlation Eq.(14)

$$\begin{aligned} \gamma &= \langle (\mathbf{n} \cdot \mathbf{n}_0) \rangle \\ &= \int (\mathbf{n} \cdot \mathbf{n}_0) F(\mathbf{n}|B_N, \mathbf{I}) w(\mathbf{B}_N|\mathbf{I}) W_I(\mathbf{I}) d^3 B_N d\Omega(\mathbf{n}) d^3 I. \end{aligned} \quad (19)$$

This gives us the long time ($t \gg T_N$) electron spin polarization (Eq.(14)). Straightforward calculation gives:

$$\begin{aligned} \gamma(x) &= \frac{2}{\pi x^3} \int_0^\infty dy \int_0^\infty dz \frac{[2y \cosh(2y) - \sinh(2y)]^2}{yz \sinh(2y)} \\ &\times \exp \left[- \left(1 + \frac{1}{x^2} \right) z^2 - \left(\frac{y}{z} \right)^2 \right], \end{aligned} \quad (20)$$

where

$$x = \frac{\langle a \rangle^2 \Delta_I^2}{\Delta_B'^2} = \frac{\langle a \rangle^2}{\langle a^2 \rangle - \langle a \rangle^2} \quad (21)$$

is the relative dispersion of the hyperfine magnetic field of the localized electron acting on nuclei and $\langle a^2 \rangle = \Delta_B^2 / \Delta_I^2$. If all the nuclei in the unit cell have the same hyperfine constant $x = N_L / (N - N_L)$.

In Fig. 1. we show γ as function of $\langle a^2 \rangle / \langle a \rangle^2 \approx N_L / N$. In the limit of large x , where the number of nuclei contributing to the field, \mathbf{B}_N , is close to the number of nuclei whose

total nuclear spin is conserved, $\gamma(x) \approx 1$. This case is realized in the "box model" and also at short times $t < T_N$. The number of nuclei that are included in the electron-nucleus spin system of the Hamiltonian Eq.(2), and whose total spin is conserved, increases with time, which in turn decreases γ . However this only holds for $t < T_{d-d}$ the dipole-dipole nuclear spin relaxation time in the dipole field of its neighbors. For spherical QDs with 10^5 nuclei confined in an infinite potential barrier (note that $N_L \approx N_{\text{tot}}/2.8$, where N_{tot} is the total number of nuclei in the QD), our approach is valid for the time interval $10T_N < t < 100T_N$ (see Appendix). The calculation in this model shows that the number of nuclei that need be included at $t \sim T_{d-d}$ is $\sim 2.1N_L$ ($\gamma \approx 0.4$) and does not reach its maximum value $N \approx 2.8N_L$.

It is important to note that non vanishing average spin polarization at time $t \gg T_N$ ($\gamma \neq 0$) means that there is a significant probability that an ensemble of electron-nucleus spin systems retains its initial spin state.

IV. SPIN DECOHERENCE IN A STRONG EXTERNAL MAGNETIC FIELD.

A strong external magnetic field, B , ($B \gg B_N$) significantly changes the process of electron spin relaxation. In this large field the Zeeman splitting of the electron spin levels is larger than their inhomogeneous broadening in the hyperfine nuclear magnetic field. The total magnetic field acting on the electron is now effectively directed along the external magnetic field. The nuclear hyperfine fields only perturb the precession frequency of the electron spin about the external magnetic field direction.

Consider, now, the effect of a strong external magnetic field on the electron spin polarization. The motion of the spin in the total magnetic field is again described by Eq. (9) where, now, $\mathbf{n} = (\mathbf{B} + \mathbf{B}_N)/|\mathbf{B} + \mathbf{B}_N|$. Averaging Eq. (9) over the ensemble, using the distribution of nuclear magnetic fields in Eq.(4), we obtain:

$$\begin{aligned} \langle \mathbf{S}(t) \rangle = & R_{\parallel}(t)(\mathbf{S}_0 \cdot \mathbf{b})\mathbf{b} + R_{\perp}^0(t)[\mathbf{S}_0 - (\mathbf{S}_0 \cdot \mathbf{b})\mathbf{b}] \\ & + R_{\perp}^1(t)[(\mathbf{S}_0 - (\mathbf{S}_0 \cdot \mathbf{b})\mathbf{b}) \times \mathbf{b}] , \end{aligned} \quad (22)$$

where

$$R_{\parallel}(t) = R_{\parallel}^{\infty} + \Delta R_{\parallel}(t) ,$$

$$R_{\perp}^0(t) = R_{\perp}^{\infty} + \Delta R_{\perp}^0(t) ,$$

and $\mathbf{b} = \mathbf{B}/B$ is a unit vector along the external magnetic field,

$$R_{\parallel}^{\infty} = \left\langle \frac{[B + (\mathbf{B}_N \cdot \mathbf{b})]^2}{|\mathbf{B} + \mathbf{B}_N|^2} \right\rangle = 1 - 2R_{\perp}^{\infty} , \quad (23)$$

and

$$\begin{aligned} R_{\perp}^{\infty} &= \frac{1}{2} \left\langle \frac{B_N^2 - (\mathbf{B}_N \cdot \mathbf{b})^2}{|\mathbf{B} + \mathbf{B}_L|^2} \right\rangle \\ &= \frac{1}{2\sqrt{\pi}} \int_{-\infty}^{\infty} dz \int_0^{\infty} dy \frac{y \exp(-z^2 - y)}{(\beta + z)^2 + y} \end{aligned} \quad (24)$$

is the value of $\langle \mathbf{S}(t) \rangle$ in the long time limit $t \gg T_{\Delta}$, and $\beta = B/\Delta_B$. The time dependent components are given by

$$\begin{aligned} \Delta R_{\parallel}(t) &= \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} dz \int_0^{\infty} dy \frac{y \exp(-z^2 - y)}{(\beta + z)^2 + y} \\ &\quad \times \cos \left[\sqrt{(\beta + z)^2 + y} \frac{t}{T_{\Delta}} \right] , \\ \Delta R_{\perp}^0(t) &= E(t) - \frac{\Delta R_{\parallel}(t)}{2} , \end{aligned} \quad (25)$$

where

$$E(t) = \exp \left[- \left(\frac{t}{2T_{\Delta}} \right)^2 \right] \left[\cos \left(\frac{\beta t}{T_{\Delta}} \right) - \frac{t}{2\beta T_{\Delta}} \sin \left(\frac{\beta t}{T_{\Delta}} \right) \right] , \quad (26)$$

and

$$\begin{aligned} R_{\perp}^1(t) &= \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} dz \int_0^{\infty} dy \frac{[(\beta + z)^3 + zy] \exp(-z^2 - y)}{[(\beta + z)^2 + y]^{3/2}} \\ &\quad \times \sin \left[\sqrt{(\beta + z)^2 + y} \frac{t}{T_{\Delta}} \right] . \end{aligned} \quad (27)$$

Equation (22) simplifies considerably for strong magnetic fields. Calculation of the coefficients in Eqs. (23,24,25) in the limit $\beta \gg 1$ gives:

$$\begin{aligned} \langle \mathbf{S}(t) \rangle &\approx \\ &\left\{ 1 - \frac{1 - \cos(\omega_B t)}{\beta^2} \exp \left[- \left(\frac{t}{2T_{\Delta}} \right)^2 \right] \right\} (\mathbf{S}_0 \cdot \mathbf{b}) \mathbf{b} \end{aligned}$$

$$\begin{aligned}
& + \left\{ \left[\cos(\omega_B t) + \frac{1 - \cos(\omega_B t)}{2\beta^2} \right] [\mathbf{S}_0 - (\mathbf{S}_0 \cdot \mathbf{b})\mathbf{b}] \right. \\
& \left. + \sin(\omega_B t)[(\mathbf{S}_0 - (\mathbf{S}_0 \cdot \mathbf{b})\mathbf{b}) \times \mathbf{b}] \right\} \exp \left[- \left(\frac{t}{2T_\Delta} \right)^2 \right] .
\end{aligned} \tag{28}$$

One can see that in strong magnetic fields, $B \gg \Delta_B$ the component of spin along \mathbf{B} is conserved, while its two transverse components precess with a frequency $\omega_B = \mu_B g_e B / \hbar$, and decay as a result of the inhomogeneous broadening of the levels in the random magnetic field of nuclei, respectively. The dephasing arises from the dispersion of the nuclear field along the external magnetic field, which leads to an inhomogeneous dispersion of the electron precession frequency. The perpendicular components of the nuclear magnetic field change the direction of the precession axis by a small angle of $\sim B_N/B$ and lead to a dephasing rate that is $(B_N/B)^2$ smaller than that due to the dispersion of the nuclear field along the external field. This result is consistent with the small ratio of the *lifetime broadening* to the *secular broadening* describing the transverse relaxation time T_2 in strong magnetic fields (see e.g. Ref. [17]).

Figure 2 shows the time dependence of the various components of $\langle \mathbf{S}(t) \rangle$ which occur in Eq.(22). The both longitudinal and transverse components of the electron spin polarization tend to a steady state value after several oscillations. The number of oscillations grows with increasing a magnetic field. Increasing the magnetic field also changes the steady state value of the longitudinal component $R_{\parallel}(\infty)$ (from 1/3 to 1) and the transverse component $R_{\perp}^0(\infty)$ (from 1/3 to 0), while the steady state value of $R_{\perp}^1(\infty)$ is zero for all values of B . In a strong magnetic field when the nuclear spin relaxation mechanism of the longitudinal spin polarization is suppressed phonons can again play an important role (see ref. [18]).

The important characteristic measured in steady state experiments, such as Hanle effect measurements (e.g., see [3]), is the average electron polarization for its lifetime, $(1/\tau) \int \langle S(t) \rangle \exp(-t/\tau) dt$, where τ is the lifetime of the localized electrons. Comparison with Eq.(22) shows that this average polarization is characterized by:

$$\rho_{\parallel}(\beta, \tau) = \frac{1}{\tau} \int_0^{\infty} R_{\parallel}(t) \exp(-t/\tau) dt ,$$

$$\rho_{\perp}^{0,1}(\beta, \tau) = \frac{1}{\tau} \int_0^{\infty} R_{\perp}^{0,1}(t) \exp(-t/\tau) dt . \quad (29)$$

The dependence of these respective terms on the magnetic field is shown in Fig. 3 for different values of the electron lifetime.

Equation (22), as is Eq.(10), is derived assuming a time independent frozen fluctuation of the nuclear field acting on the electrons. In zero external magnetic field, this no longer holds at longer times such that we must take into account the nuclear spin precession in the inhomogeneous hyperfine field of the electron. Nuclear spins precessing at different rates about the average electron spin direction create a time dependent hyperfine field with components perpendicular to the original direction of the frozen fluctuation. As a result the average electron spin projection follows the new direction of the slowly varying nuclei field. The characteristic time of this slow process is determined by the dispersion of the nuclear spin precession frequency in the inhomogeneous field of the electron $T_N^{-1} \approx (\mu_B g_e / \hbar) \sqrt{\langle a^2 \rangle - \langle a \rangle^2} \sim (\mu_B g_e / \hbar) \sqrt{\langle a^2 \rangle}$. This second regime of spin relaxation begins when $t \geq T_N$.

In a strong external magnetic field $B \gg \Delta_B$, the average electron spin is directed along this strong field, independent of the nuclear hyperfine fields ($B_N \ll B$). Although the nuclei precess with different frequencies in the inhomogeneous electron field, the electron is effected only by the component of the nuclear field along the external field. As a result the nuclear magnetic field acting on the electron spin is frozen for times much longer than T_N . Thus a frozen fluctuation model of the nuclear hyperfine field is valid when describing the dephasing dynamics of the electron spin polarization in an ensemble of quantum dots in strong magnetic fields. As we mentioned above this consideration is limited by a low enough temperature and the time scale of the nuclear dipole-dipole interaction.

In each QD, the motion of the electron spin in the hyperfine field of the frozen fluctuation of the nuclei is coherent. The dephasing is a result of inhomogeneous broadening of the electron spin levels in the ensemble of quantum dots. This makes it possible to recover the transverse electron spin polarization using the spin echo technique [12], which also can be

used for quantum computation [19].

V. DISCUSSION

We have determined the time dependence of the electron spin relaxation rate arising from its interaction with nuclear spins for an ensemble of QDs, or equivalently, averaged over a large number of successive measurements of a single dot. This gives us the value of T_2^* usually measured in ensemble experiments.

The electron spin relaxation time was studied in three types of QDs: GaAs natural quantum dots, which are island-like dots formed by a fluctuation of the GaAs quantum well thickness [20]; self organized InAs QDs [21], and nanocrystal CdSe QDs [22]. We can estimate the value of T_Δ for each of these dots. In GaAs natural QDs, using values of A^j from Ref. [13] and Eq.(6), and taking the dot to contain 10^5 nuclei, Eq.(11) gives $T_\Delta \sim 1.0$ ns. The values of A^j are not experimentally determined for InAs. Assuming that compound ionicity does not significantly vary among the semiconductors GaAs, InSb and InAs [13,23], we take the hyperfine constants, A^j , for As and In ions from [13] and [24]: $A_{As}^j = 47\mu\text{eV}$ and $A_{In}^j = 56\mu\text{eV}$, respectively. The large value of the In nuclear spin, $I = 9/2$, strongly effects the magnitude of the hyperfine interaction in InAs, and shortens the spin dephasing time. To estimate this time we need to know the number of nuclei in the electron localization volume. In the self organized QDs, where the electron wave function strongly depends on the QD shape and barrier height, we used the geometrical volume of the dot to determine the number of nuclei, N_L , in Eq.(7). The QDs studied in [21] contain 1000-4000 nuclei [25]. This leads to electron spin relaxation times on the order of 50-100 ps, which is close to the value of gT_2^* measured in these dots. The hyperfine interaction constants in CdSe are also not experimentally determined. In this material only a fraction of the nuclei (25 % of the Cd ions) have a magnetic moment and these have spin $I = 1/2$. As a result, the electron spin interaction with the nuclei in these QDs is weak. We shall take for the hyperfine constant in CdSe half that of Cd^{111} [26], assuming completely covalent bonding

in this material. This leads to $A_{Cd}^j = 12\mu\text{eV}$. Taking the CdSe lattice constant $a_0 \approx 4.2\text{\AA}$ [27] we calculate $N_L = 3,500$ for the ground 1S electron state in spherical nanocrystals with a radius 28\AA . Only 1/8 of these nuclei contribute to dephasing. This leads to $T_\Delta \approx 1.6\text{ ns}$, which is consistent with the experimental measurement $T_2^* = 2.5\text{ ns}$ [22]

In conclusion, we developed a theory of electron spin relaxation in QDs arising from their hyperfine interaction with the QD nuclei. The relaxation is determined by three physical processes: (i) – the precession of the electron spin in the hyperfine field of the frozen fluctuation of the nuclear spins; (ii) – the precession of the nuclear spins in the hyperfine field of the electron; and (iii) – the precession of a nuclear spin in the dipole field of its neighbors. These processes have three disparate characteristic times. For GaAs QDs with 10^5 nuclei, they are $\sim 1\text{ ns}$, $\sim 300\text{ ns}$, and $\sim 100\mu\text{s}$, respectively. The last of these times is so long that many other electron spin relaxation mechanisms can be more important on this time scale. An external magnetic field suppresses the relaxation of the spin component along the magnetic field. The transverse components of the electron spin polarization relax completely in a time on the order of the electron precession period in the field of the frozen nuclear fluctuation. Comparison with experimental data shows that the hyperfine interaction with nuclei is the dominant mechanism of electron spin relaxation in QDs.

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APPENDIX: THE NUMBER OF NUCLEI INTERACTING WITH A LOCALIZED ELECTRON IN A QD

The number of nuclei, N , "interacting" with a localized electron and forming an electron-nuclear system in thermodynamic equilibrium is generally a time dependent quantity. Formally, the electron interacts at all times with all those nuclei of the crystal lattice sites where

its wave function is nonzero. However, the strength of this interaction decreases with the distance from the QD center as the square of the electron wave function, and the random magnetic field \mathbf{B}_N , which determines the electron spin precession is determined only by the relatively small number of nuclei, N_L , in the "effective" volume of the electron localization V_L (see Eq.8). This volume (or alternatively N_L) completely determines the electron spin relaxation at short times $t < T_N$ during which nuclear spins, and, therefore, the corresponding field \mathbf{B}_N do not change their directions.

The precession of the nuclear spins in the hyperfine field of the electron causes time dependent changes in \mathbf{B}_N . This time dependent $\mathbf{B}_N(t)$ in each dot is determined by a macroscopic number of initial conditions. The correlation $\langle \mathbf{B}_N(t) \mathbf{B}_N(0) \rangle$ (see Eq.14) is now an important characteristic of an ensemble of quantum dots. The ensemble of localized electrons completely loses its memory of the initial spin polarization if this correlation is zero. Although $\langle \mathbf{B}_N(t) \mathbf{B}_N(0) \rangle$ can be calculated for any time, t , in principle, in the long time limit $t \gg T_N$, when the values of $\mathbf{B}_N(t)$ are randomly distributed, one can find this correlation using statistical physics techniques.

Let us separate the nuclei interacting with a localized electron during a time t into two groups: (1) those nuclei whose spin rotates through an angle $\omega_i t \geq 1$ and (2) those whose spin rotates through an angle $\omega_i t \leq 1$, where ω_i is the precession frequency of the nuclear spin in the hyperfine field of the localized electron. The second group can be considered as nuclei that "do not interact" with the electron. For times $t \gg T_N$ the first group automatically includes the N_L nuclei that determines the field \mathbf{B}_N . The number of nuclei $N(t)$ "interacting" with the electron, and thus belong to the first group, increases with time and determines $\mathbf{B}_N(t)$, whose variation is restricted by conservation of the total spin of the electron-nuclear spin system. Randomness of the distribution of the $N(t)$ nuclear spins limited by the conservation of B_N and \mathbf{I}_Σ is the one major assumption of this statistical model.

For nuclei that interact strongly through the "indirect" hyperfine interaction, i.e. nuclei whose spins rotate through an angle $\omega_i t \gg 1$ in the electron hyperfine field, this assumption

is justified. The error in the statistical approach is connected with the border nuclei for which $\omega_i t \sim 1$. These nuclei strongly interact with the electron, but, on the other hand, they still "remember" their initial direction at $t = 0$. The number of such nuclei is on the order of $\Delta N \approx t \partial N / \partial t$. As a result the relative error of our estimate is $\delta N = \Delta N(t) / N(t) \approx (t/N) \partial N / \partial t$.

The time dependence of $N(t)/N_L$ and δN in spherical QDs with an infinite barrier is shown in Fig.4. The time is measured in the unit of $T_N^0 \sim (32/\pi)(\hbar/A)(a^3/v_0)$, the nuclear precession time at the center of the QD, where a is the QD radius. In GaAs QDs with 10^5 nuclei, this time is on the order of microseconds. One sees that $N < N_L$ for $t < 10T_N^0$. In this time period $\delta N \geq 1$ and our statistical approach is not valid. However $N(t)$ increases and $\delta N(t)$ decreases rapidly with time, and $\delta N \sim 0.25$ at $t \approx 20T_N^0$. After this time the nuclei which give the main contribution to the magnetic field, B_N , acting on the electron are in statistical equilibrium. This allows us to use Eq.(20).

REFERENCES

- [1] J. K. Kikkawa and D. D. Awschalom, Phys. Rev. Lett. **80**, 4313 (1998); Nature (London) **397**, 139 (1999).
- [2] D. Loss and D. P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).
- [3] *Optical Orientation*, Eds. B. Meier and B. P. Zakharchenya, Ch.III, North Holland, Amsterdam, 1984
- [4] A. Khaetskii and Yu. V. Nazarov. Phys. Rev. B **61**, 12639 (2000).
- [5] R. I. Dzhiyev, B. P. Zakharchenya, E. L. Ivchenko, V. L. Korenev, Yu. G. Kusraev, N. N. Ledentsov, V. M. Ustinov, A. E. Zhukov, and A. F. Tsatsl'nikov, JETP Lett. **65**, 804 (1997).
- [6] H. Nickolaus, H. J. Wunsche, F. Henneberger, Phys. Rev. Lett. **81**, 2586 (1998).
- [7] M. Z. Maialle, Phys. Rev. B **61**, 10877 (2000).
- [8] A. Abragam and B. Bleaney, *Electron Paramagnetic Resonance of Transition Atoms* Oxford University Press, London/New York, 1970.
- [9] T. Takagahara, Phys.Rev. B **62**, 16840 (2000).
- [10] G. Lampel, Phys. Rev. Lett. **20**, 491 (1968).
- [11] *Optical Orientation*. Eds. B. Meier and B. P. Zakharchenia, Ch.II, ChVI, Ch X, North Holland, Amsterdam, 1984
- [12] A. Abragam, *Principles of Nuclear Magnetism* Clarendon Press, Oxford, p.172, 1996.
- [13] D. Paget, G. Lampel, B. Sapoval, and V. I. Safarov, Phys. Rev. B **15**, 5780 (1977).
- [14] Spin conservation can be destroyed by nuclear dipole-dipole interaction effects or by spin diffusion.
- [15] S. M. Raybchenko and U. G. Semenov, Sov. Phys. JETP **84**, 1412 (1983).

- [16] L. D. Landau and E. M. Lifshitz, *Statistical Physics*, Pergamon Press, Oxford, 1976.
- [17] C. P. Slichter, *Principles of Magnetic Resonance*, 2nd edition, Springer-Verlag, Berlin Heidelberg, New York, 1980, pp.180-182.
- [18] S. I. Erlingsson, Yu. V. Nazarov, and V. I. Fal'ko, Phys. Rev. B **64**, 195306 (2001).
- [19] N. Gershenfeld and I. L. Chuang, Science **275** 350 (1997).
- [20] D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park, Phys. Rev. Lett. **76**, 3005 (1996).
- [21] R. Epstein, D. T Fuchs, W. V. Schoenfeld P. M. Petroff, and D. D. Awschalom, Appl. Phys, Lett. **76**, 733 (2001).
- [22] J. A. Gupta, D. D. Awschalom, X. Peng, and A. P. Alivisatos, Phys. Rev. B **59** R10421 (1999).
- [23] J. C. Phillips, *Bonds and Bands in Semiconductors*, Academic Press, NY, 1973.
- [24] M. Gueron, Phys. Rev. **135**, A200 (1964).
- [25] P. M. Petroff, private communication.
- [26] CdSe, A. K. Kohn and D. J. Miller, *Atomic data and nuclear data tables*, **33**, 235-253, Academic Press, (1985).
- [27] A. G. Milnes and D. L. Feucht, *Heterojunctions and Metal-Semiconductor Junctions*, Academic Press, New York -London, 1972.

FIGURES

FIG. 1. The dependence of the ensemble averaged electron spin polarization on the ratio of dispersion of the electron hyperfine field acting on the nuclei to its ensemble averaged value.

FIG. 2. The time dependence of the longitudinal, (*a*), and transverse, (*b*) and (*c*), components of the ensemble averaged electron spin polarization calculated for different magnetic fields. The curves are calculated for $\beta = 0, 1, 2, 3, 5$ respectively.

FIG. 3. The magnetic field dependence of the longitudinal, (*a*), and transverse, (*b*) and (*c*), components of the steady state electron spin polarization. Calculations are done for electron lifetimes $\tau/T_\Delta = 0, 0.2, 0.4, 0.8, 1.4, 3.5$, and 10.

FIG. 4. Time dependence of the ratio of the number of nuclei, $N(t)$, "interacting" with an electron, in a spherical QD with an infinite potential barrier, to N_L and the relative error, $\delta N(t)$.







